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A Proportional Counter for Mössbauer Spectroscopy by Scattered Electrons

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A proportional counter with a very small sensitive volume ($250\,mm^2 \times 3\,mm$) has been constructed for Mössbauer spectroscopy by scattered electrons. More than 20 % effect with a 100 % baseline has been obtained for an unenriched sample of Type 310 stainless steel with a helium-10 % methane mixture flowing through the counter. Some features of the counter are described.

Mössbauer experiment has usually been performed by transmission geometry, in which the recoil-less γ rays from a source are counted after passing through an absorber. It is also possible to observe the Mössbauer effect by detecting the resonantly reemitted γ rays, the electrons resulting from the internal conversion competitive with the γ-ray emission, the X rays or Auger electrons accompanying the conversion process. Some workers have reported successful Mössbauer measurements by reemitted γ rays or K-conversion X rays in the backscattering geometry.[1~12] However, the technique of the Mössbauer measurement by scattered electrons (conversion electrons and Auger electrons) has not been established because of the difficulties in the efficient detection of low energy electrons.

For the ^{57}Fe Mössbauer measurement, 7.3-keV K-conversion electrons from the 14.4-keV level and about 6-keV Auger electrons accompanying the conversion process

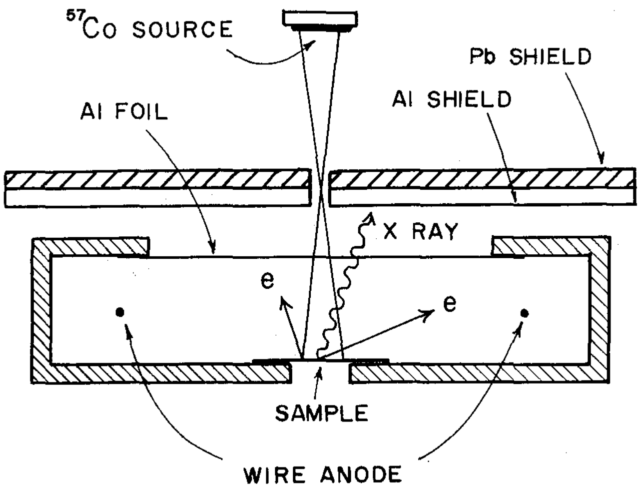

Fig. 1. Proportional counter with 2π-backscatterer geometry by Swanson and Spijkerman.

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are available. Since the range of these low energy electrons in metal is very small (~3000 Å), the Mössbauer spectroscopy by the scattered electrons is a favorable tool to study the upper-most surface layer of a sample.

A proportional counter for detecting the scattered electrons was designed by Swanson and Spijkerman.[13,14] As shown in Fig. 1, the anode of their counter consists of two stainless steel wires. The collimated incident radiation was directed through the center region between the two anode wires in order to make the counter less sensitive for the incident radiation. Samples to be studied were placed inside the counter at the wall opposite to the window for the incident radiation. In such a 2π-backscatterer geometry, 2~3 % Mössbauer effect for an iron foil was obtained without an X-ray interference by flowing a helium-10 % methane mixture through the counter. They could also operate the counter as a resonance detector with a 50 %-enriched stainless steel foil inside it.[15]

Recently, the technique has been applied to study on corrosion products formed on a surface of iron film by Onodera *et al.*[16] Krakowski and Miller have theoretically discussed resonance line width, resonance area, and magnitude of the effect for Mös-

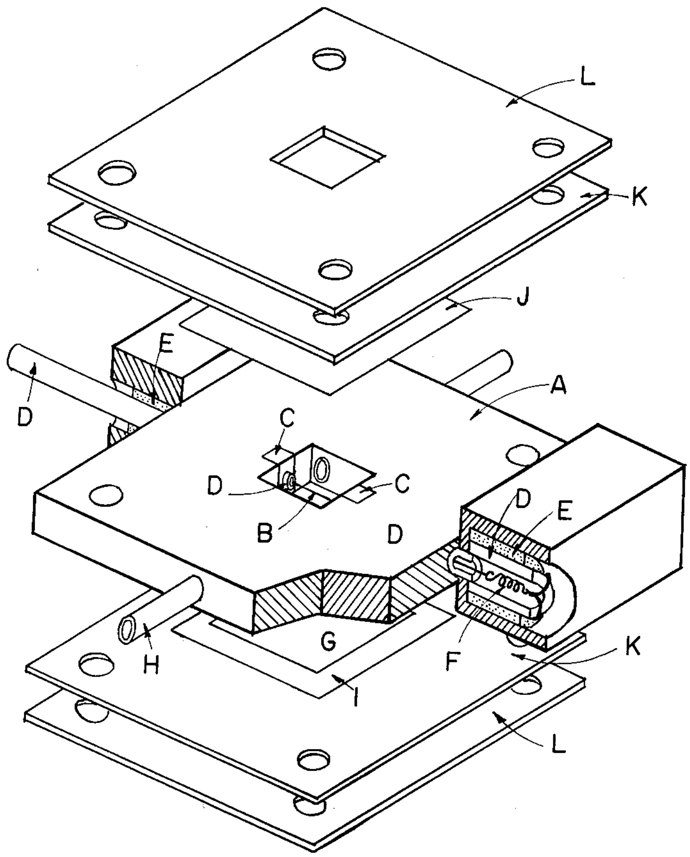

Fig. 2. Proportional counter for the Mössbauer experiment by scattered electrons: A, counter frame (Lucite); B, anode wire (tungsten); C, Teflon; D, stainless steel pipe; E, Teflon pipe; F, steel spring; G, sample foil; H, gas inlet; I, aluminium foil; J, alumium-evaporated Mylar foil; K, rubber sheet; L, brass plate.

sbauer backscattering spectra by the scattered electrons.[17]

Based in part on the work by Swanson and Spijkerman,[13] we have designed a counter further optimized for the scattered electrons. As shown in Fig. 2, our counter mainly consists of a 3-mm thick Lucite frame with a square hole (16×16 mm) at the center. Aluminium was evaporated on the surface of the plate to make it electrically conductive. One side of the hole was covered with a thin aluminium-evaporated Mylar film (~ 1 mg/cm^2), while the other was covered with a sample foil. In order to avoid the leakage of counter gas, 1-mm thick rubber sheets were sandwiched between the counter frame and brass plates, which were fixed together tightly, as shown in Fig. 2.

As the anode of our counter only one wire is used instead of two. Since the detection efficiency of our counter of a very small sensitive volume (250 mm$^2 \times 3$ mm) is practically zero for the incident radiation, there is no need to use a counter with two anode wires. In fact, a 3-mm thick helium gas layer at 1 atm has efficiencies of less than 0.01 % for 6.3-keV X rays and of less than 0.001 % for the 14.4-keV γ rays. The anode wire of a 50-μm diam tungsten was stretched by a small steel spring. The wire was covered with 1.5-mm diam stainless steel pipes at both the ends to avoid the electric discharge due to a large field gradient. Teflon pipes of 4-mm diam were used as insulators between the stainless steel pipes and the counter frame. During the operation of the counter, the flow of a commercial gas mixture (90 % He+10 % methane) was maintained at a rate of about 1 ml/min.

As a Mössbauer source, we used ^{57}Co of about 7 mCi diffused in copper. A 5-mm thick lead plate with a 6-mm diam hole restricted the radiation to the center area of a sample. In order to avoid the emission of unfavorable photoelectrons from the sample foil by the 6.3-keV X rays, a 5-mm Lucite plate was placed between the counter and the source, which could absorb more than 99 % of these X rays. The reduction of the

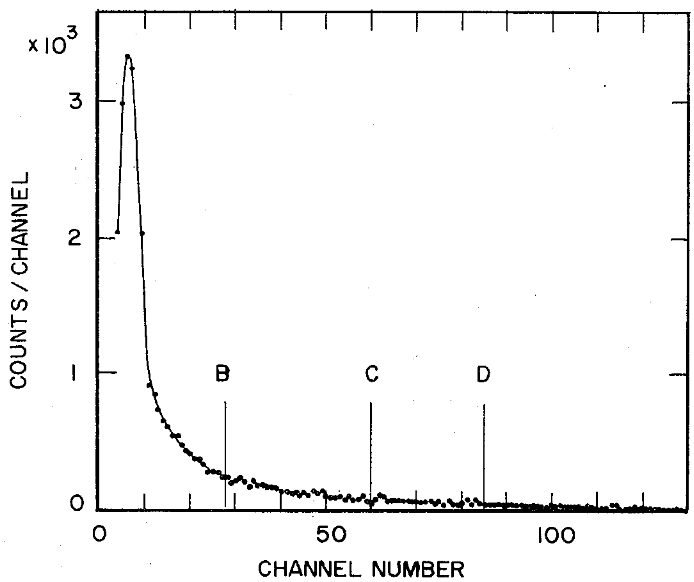

Fig. 3. Typical energy spectrum of electrons scattered from a sample of Type 310 stainless steel.

14.4-keV γ-ray intensity by the absorber was about 50%. As a standard sample, an unenriched stainless steel foil of Type 310 (125 μm thick) was used because of its simple Mössbauer spectrum. When using an iron foil, the Mössbauer spectrum was changing gradually for a long measuring period probably owing to the surface oxidation.

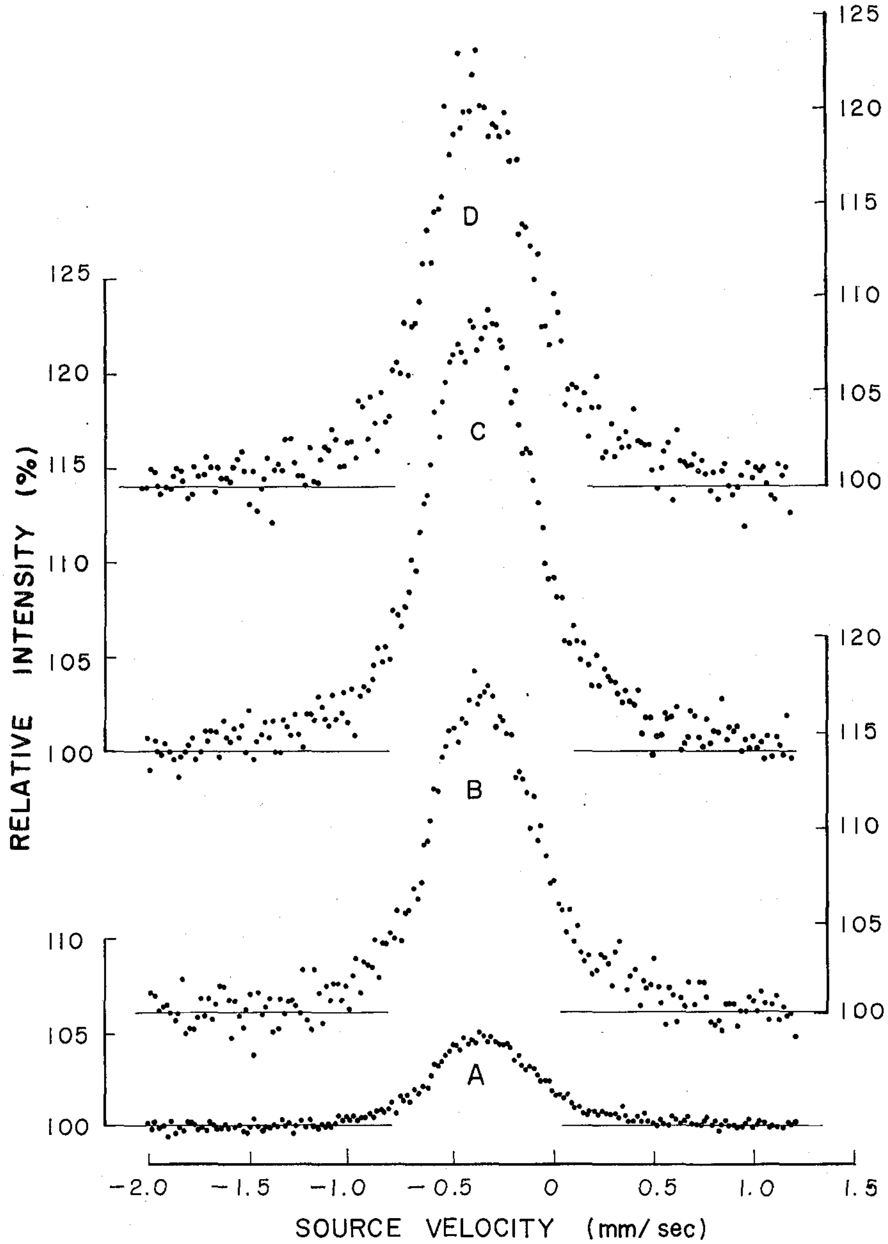

Fig. 4. Mössbauer spectra for Type 310 stainless steel. Spectrum A was obtained by counting all the signals from the counter. Spectra B, C, and D were obtained by setting the lower discriminator to energy levels B, C, and D indicated in the figure a, respectively.

Before the Mössbauer measurement, our counter was tested with various voltages applied to the anode wire. The counter could be operated in the proportional region at the anode potential ranging from 500 V to 800 V, while above 900 V it behaved as a G–M counter. Applying a voltage more than 700 V, we could easily separate the electron signals from the noise level. During the Mössbauer measurement the anode was kept at a potential of about 700 V. A typical energy spectrum of the electrons scattered from the stainless steel sample is shown in Fig. 3.

The Mössbauer spectrometer used in the present work consists of commercially available units of a function generator, a transducer driving unit and a linear velocity transducer (Elscint Ltd.). We obtained about 4.4 %-Mössbauer effect by counting all signals from the counter. We also measured a Mössbauer spectrum by a counter of the same type, but with a much larger sensitive volume ($25\,cm^2 \times 1\,cm$). Only 2.0 % effect was obtained by this larger counter. This result implies that, for an efficient detection of the electrons scattered from a sample, a smaller counter may be more advantageous because of its less sensitivity for the incident radiation or the electrons from surroundings such as counter wall, window material and lead collimator.

In Fig. 4 Mössbauer spectra obtained by setting a lower discriminator at various points in the energy spectrum of Fig. 3 are shown. It should be noted that a proper setting of the discriminator enhances the effect remarkably. The background of the spectra is caused mainly by 7.3-keV photoelectrons due to the 14.4-keV γ rays and by about 100-keV electrons due to the photoeffect or the Compton scattering of the 122-keV γ rays. A low energy electron can deposit about 4 keV of energy in the 3-mm thick helium gas layer, while a 100-keV electron deposits only less than 0.5 keV in the same layer. Because of this fact, unfavorable signals due to high energy electrons are easily eliminated by a proper discriminator setting. On the contrary, the 7.3-keV photoelectrons ejected by the 14.4-keV γ rays cannot be separated from the electrons accompanying the Mössbauer resonant absorption. According to the calculation by Spijkerman and Travis,[15] the number of conversion electrons associated with the Mössbauer absorption is about ten times larger than that of photoelectrons due to the 14.4-keV γ rays for a pure ^{57}Fe samples. The present result of about 20 %-Mössbauer effect for the unenriched stainless steel foil approximately agrees with their calculation.

The present work strongly suggests that a counter with a smaller sensitive layer is more advantageous for the Mössbauer experiment. Tests with a 1-mm thick counter are now in progress.

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